Heterogeneity of atmospheric ammonia at the landscape scale and consequences for environmental impact assessment

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A B S T R A C T

We examined the consequences of the spatial heterogeneity of atmospheric ammonia (NH_3) by measuring and modelling NH_3 concentrations and deposition at 25 m grid resolution for a rural landscape containing intensive poultry farming, agricultural grassland, woodland and moorland. The emission pattern gave rise to a high spatial variability of modelled mean annual NH_3 concentrations and dry deposition. Largest impacts were predicted for woodland patches located within the agricultural area, while larger moorland areas were at low risk, due to atmospheric dispersion, prevailing wind direction and low NH_3 background. These high resolution spatial details are lost in national scale estimates at 1 km resolution due to less detailed emission input maps. The results demonstrate how the spatial arrangement of sources and sinks is critical to defining the NH_3 risk to semi-natural ecosystems. These spatial relationships provide the foundation for local spatial planning approaches to reduce environmental impacts of atmospheric NH_3.

Keywords:
Ammonia
Critical level
Landscape scale
Dispersion modelling
Spatial planning

1. Introduction

Most atmospheric ammonia (NH_3) originates from agricultural activities (Misselbrook et al., 2000; Van der Hoek, 1998). Intensive livestock farming, i.e. large pig and poultry units, represent substantial NH_3 point sources, due to their high stocking density. Other NH_3 sources include biomass burning, fuel combustion and industrial processes such as the production of nitrogen (N) fertilisers (Bouwman et al., 1997). High atmospheric NH_3 concentrations are directly toxic to plants through stomatal uptake as soon as the uptake exceeds the detoxification capacity (Fangmeier et al., 1994). Ammonia deposition (and deposition of other forms of reactive N) can lead to eutrophication and acidification of sensitive ecosystems, causing changes in biodiversity (Cape et al., 2009b; Cellier et al., 2009; Krupa, 2003; Pitcairn et al., 2009). Studies have been conducted to quantify the effect of NH_3 emission sources on surrounding ecosystems, e.g. Fowler et al. (1998) quantified concentrations and deposition fluxes within 300 m of a poultry farm in Scotland using measurements and deposition modelling, showing rapidly decreasing concentrations with distance from the source. Pitcairn et al. (1998, 2002) analysed the impact of such deposition fluxes on woodland flora and Frati et al. (2007) studied the effect of pig farm emissions on sensitive vegetation (lichens). Sutton et al. (1998) compared deposition estimates based on different scales, ranging from field to landscape to national scale and concluded that, due to the spatial variability of NH_3, the quality of an environmental impact assessment is dependent on the spatial resolution of the deposition data used. Dragosits et al. (2002) provide a more detailed analysis of the landscape study in Sutton et al. (1998): Emission, transport and deposition were modelled within a 5 km $\times$ 5 km landscape in England at a 50 m grid resolution; however, no NH_3 measurements were made to verify the estimates. Theobald et al. (2001) and Dragosits et al. (2006) focused on strategies to reduce the effect of emission hotspots on ecosystems by locating tree belts around the sources, indicating the importance of relative spatial location of sources and sinks, and assessed possible landscape planning measures to decrease potential effects on sensitive habitats.

As an approach to assess the risk of environmental impacts by air pollutants, the United Nations Economic Commission for Europe (UNECE) has developed critical thresholds of pollutant concentrations and deposition fluxes: Critical Levels (CLEs) and Critical Loads (CLs). A CLE is a pollutant concentration in the atmosphere above which plants or ecosystems may be directly negatively affected (Posthumus, 1988). Recently, long term CLEs of NH_3 were reviewed and new, lower values proposed and adopted by the UNECE (Cape et al., 2009a; Sutton et al., 2009a; UNECE, 2007): 1 $\mu g\ NH_3\ m^{-3}$ for the most sensitive ecosystems, i.e. where lichens and bryophytes are part of the ecosystem integrity, and $3 \pm 1\ \mu g\ NH_3\ m^{-3}$ for higher plants in other semi-natural ecosystems. A CL is a pollutant deposition below which no significant harmful effects on the environment are expected to occur according to current knowledge (Posthumus, 1988). Nitrogen (N) CLs have been defined for specific ecosystem types (see UNECE, 2010 for most up-to-date CLs). In contrast to the CLE approach, which is specifically defined for gases such as NH_3, the CL integrates all forms of reactive N and therefore requires estimates of total N deposition. According to Sutton et al. (2009b) these N deposition estimates are inherently more uncertain, and for assessing the environmental risk imposed by NH_3, it is much easier to measure NH_3 concentrations and examine exceedance of the CLE than to verify CL exceedances by measurement. However, until the recent revision of CLEs, exceedance of CLs has been more commonly used for impact assessments of atmospheric N. For atmospheric NH_3, this may reflect that previous long-term NH_3 CLEs were set at much less precautious level than associated values of N CLs (e.g. Burkhardt et al., 1998), which was one reason for the revision of new long term NH_3 CLEs (Sutton et al., 2009b).

For assessing the environmental impact of NH_3 concentrations and deposition by modelling, it is essential to estimate NH_3 emissions accurately (Dragosits et al., 2002; Hellsten et al., 2008). Hallsworth et al. (2010) highlighted the problem of modelling NH_3 dispersion at relatively coarse scales, such as 5 km resolution, due to the high spatial variability of NH_3 emissions and showed that 5 km modelling underestimated the impact of NH_3 concentrations on semi-natural areas close to intensive agricultural areas. However, at UK national scale, standard assessments of the impact of N deposition are based on 5 km resolution modelling (Dore et al., 2007; Matejko et al., 2009). Dore et al. (2012) compared CL exceedances in the UK using data at 1 km and 5 km resolution. In contrast to results of Hallsworth et al. (2010) for CLEs, CL exceedances were not highly sensitive to grid resolution. This was attributed to the contribution of N wet deposition (which shows less local variability than dry deposition) and that all ecosystem types were included (not only nature reserves protected under the Habitat Directive, as in Hallsworth et al., 2010). However, for an individual nature reserve located a few kilometres from a major road, the standard 5 km grid data were inadequate to accurately assess local N deposition (Dore et al., 2012).

This study provides a contribution to the landscape scale analysis conducted across Europe within the NitroEurope Integrated Project (NEU) (Sutton et al., 2007), in which a landscape is defined as a spatially heterogeneous area covering several square kilometres and contains interacting ecosystems (Forman and Godron, 1981). In rural landscapes, anthropogenic processes in the form of farm management determine to a large extent N dynamics and much of its environmental impact within the landscape (Cellier et al., 2011). The NEU landscape analysis aimed to quantify N flows at the landscape scale using measurement and modelling approaches. In this study, we analysed NH_3 dispersion and its environmental impact in a 6 km $\times$ 6 km rural landscape in southern Scotland. The landscape has a diverse emission pattern with a large number of NH_3 emission hotspots, and large areas of sensitive ecosystems as potential sinks. A detailed landscape inventory of all farms and fields at field-level resolution was conducted to coincide with an intensive spatial monitoring programme of NH_3 concentrations. Ammonia dispersion and deposition was modelled at a 25 m resolution, and the environmental impact of the local NH_3 sources assessed and compared with national 1 km resolution estimates (Hallsworth et al., 2010). The results have general implications for the sustainable management of landscapes that combine both intensive livestock agriculture and ecosystems of relevance for environmental protection.

2. Site and methods

2.1. Study area

The study landscape is situated in southern Scotland, which has a temperate climate, with an annual mean temperature of ~8 °C, a typical rainfall of ~1000 mm and predominant southwesterly winds. The 6 km $\times$ 6 km area (Fig. 1) is dominated by agricultural grassland (48%), followed by moorland (21%), rough grass (13%) and woodland (10%). The moorland area with low NH_3 emissions is in the northwestern part of the landscape and is partially grazed by sheep at very low stocking densities, partly legally protected as a Site of Special Scientific Interest (SSSI), with another part undergoing peat cutting. The southeastern part is dominated by agricultural land, such as sheep and beef cattle pastures interspersed with poultry farming houses containing nearly 1.5 million laying hens. Most of the layers are kept in cage systems with manure removal by belt systems two to three times per week (farm locations circled in black in Fig. 1, with other livestock houses shaded in black). However, a number of the houses have deep-pit systems, and in most of them layers are managed as free range birds.

2.2. Landscape inventory and emissions

Detailed land cover/land use and farm activity data were obtained by a local survey carried out by Scotland's Rural College (SRUC) and the Centre for Ecology & Hydrology (CEH). Management activities were recorded for each farm building and agricultural field throughout 2008, including type and numbers of livestock housed and grazed, manure management, ventilation type and emission height, crop type and the application of mineral and organic fertiliser. Land cover/land use and farm activity data were processed with a Geographical Information System (ESRI, ArcGIS) and emissions calculated for each individual field and livestock house. Field emissions were calculated by applying UK average emission factors (EFs) of the UK NH_3 emission inventory to applications of mineral and organic fertiliser and to excreta of grazing livestock (Misselbrook et al., 2009), using fertiliser application rates provided by the farmers. Typical N contents were applied to the different types of organic fertiliser (Defra, 2010). Grazing excreta were calculated using grazing records and daily N excretion rates of the specific type of animal (Misselbrook et al., 2009). Similarly, average NH_3 EFs of the UK emission inventory were applied to calculate housing and manure storage emissions, with housing records on systems and durations provided by the farmers. However, after initial analyses, housing EFs were partly adjusted to account for specific local management practices (see Section 3.3). All spatial datasets were converted to a 25 m grid resolution for atmospheric dispersion modelling (see Section 2.4).

2.3. Spatial NH_3 concentration measurements

Monthly average concentrations were measured from April 2007 to December 2008 at 31 locations using ALPHA passive diffusion samplers (Tang et al., 2001) at a sampling height of 1.5 m above ground. Measurement locations were distributed across the study area in collaboration with farmers and landowners in the landscape. Sites were selected to cover NH_3 concentrations over different land cover types and farms. More sites were placed in NH_3 emitting areas to capture concentration gradients around emission hotspots and diffuse sources, taking the main wind direction into account. The nearest site to an emission hotspot was located 70 m downwind of a poultry house to avoid saturation of the samplers. To assess measurement precision and uncertainty, samplers were exposed in triplicate at each location. The sampling rate of the ALPHA samplers was calibrated against the DELTA denuder reference system (Sutton et al., 2001b), using the UK National Ammonia Monitoring Network methodology (NAMN, Sutton et al., 2001a). ALPHA samplers were stored in a cold room (4 °C) until analysis in the laboratory with an AMFIA NH_3 flow injection analyser, based on analysis by selective ion membrane transfer and subsequent conductivity measurement (Wyers et al., 1993).

2.4. Atmospheric dispersion and deposition modelling

Atmospheric dispersion and dry deposition of NH_3 within the study landscape was simulated using the LADD (Local Area Dispersion and Deposition) model (Hill, 1998). Loubet et al. (2009) recently reviewed LADD and other models available for simulating NH_3 dispersion. The advantages of LADD are that it operates in 3D (with

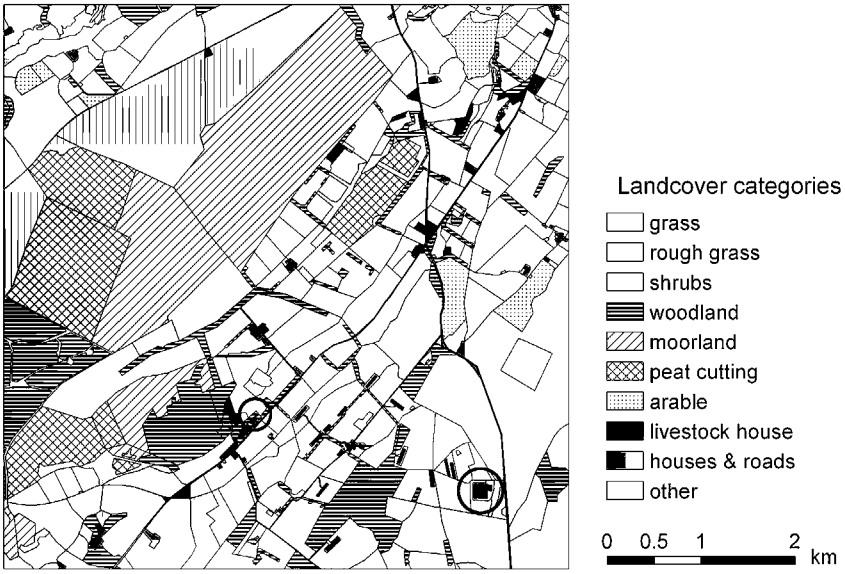

Fig. 1. Land cover types in the 6 km × 6 km study landscape in southern Scotland. The circles highlight poultry houses where manure is cleared at least twice per week. At other animal houses manure is removed less frequently (see Section 3.3).

the atmosphere represented by 44 vertical layers), is computationally fast, and accounts for land cover-specific dispersion and deposition characteristics (Loubet et al., 2009). Input data include land cover and emission data for each grid square (see Section 2.2), wind statistics and NH_3 concentrations at the domain boundaries. Suitable roughness length (z_0) and canopy resistance (R_c) for each given land cover type were selected and assigned in LADD. The roughness length is used to calculate vertical dispersion and dry deposition rates, while R_c is used in the calculation of dry deposition velocities within each grid square. Wind statistics were calculated from data collected for 30-min-intervals during 2008 at a continuous measurement site near the centre of the study area (M. Coyle, CEH, pers. comm. 2010; n.b. exact coordinates not presented for farm data confidentiality purposes). The influence of emission sources outside of the modelling domain was incorporated by setting the atmospheric concentrations for the 44 model layers at the four domain boundaries to values taken from the national FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) model run for 2008 at a 5 km × 5 km resolution (Dore et al., 2007). These boundary concentrations were highest at ground level, ranging from 1.34 μg NH_3 m^{-3} at the eastern boundary to 1.85 μg NH_3 m^{-3} in the south. The FRAME simulations used annual precipitation data for 2008 from the UK Met Office precipitation monitoring network and wind direction frequency data generated from radiosonde data for 2008.

LADD was applied for the year 2008 at 25 m × 25 m grid resolution over an area of 7 km × 7 km with the model domain extended by 500 m on all sides to limit possible edge effects. Annual average NH_3 concentrations at 1.5 m height above ground level and dry deposition were simulated and subsequently analysed with ArcGIS (ESRI).

2.5. Assessment of model performance

Model performance was assessed by comparing modelled with measured annual concentrations at the 31 sampling sites. Statistical metrics used for model evaluation were the fraction of modelled concentrations within a factor of two of observed concentrations (FAC2), the correlation coefficient (R), the geometric mean bias (MG) and the geometric variance (VG) (Chang and Hanna, 2004; Theobald et al., 2009).

$$\mathrm{FAC2} = \text{fraction of data that satisfy } 0.5 \le C_m/C_o \le 2.0 \quad (1)$$

$$R = \frac{\overline{\left(C_o - \overline{C_o}\right)\left(C_m - \overline{C_m}\right)}}{\sigma_{C_o}\sigma_{C_m}} \quad (2)$$

$$\mathrm{MG} = \exp\left(\overline{\ln C_o} - \overline{\ln C_m}\right) \quad (3)$$

$$\mathrm{VG} = \exp\left[\overline{(\ln C_o - \ln C_m)^2}\right] \quad (4)$$

where C_o are the observed (measured) concentrations, C_m are modelled concentrations, σ is the standard deviation and overlined variables represent mean values. Model performance is considered "acceptable" if FAC2 is 50% or greater, i.e. if FAC2 ≥ 0.5. MG measures the mean relative bias and only indicates systematic errors. It represents the ratio of the geometric mean of C_o to the geometric mean of C_m, thus the optimum value is MG = 1. An "acceptable" model performance is expected to result in a mean relative bias within ±30%, i.e. 0.7 < MG < 1.3. VG is a measure of mean relative scatter of a log-normal distribution and reflects both systematic and random error. The optimum value is VG = 1. An "acceptable" model would be expected to have a relative scatter of less than a factor of two (i.e. VG < 1.6) or three (i.e. VG < 3.3). Overall model performance is evaluated as acceptable when more than 50% of the criteria are met (Hanna and Chang, 2010).

2.6. Assessment of potential environmental impacts

Landscape areas with exceeded CLEs and CLs were identified to assess the risk of environmental impact on ecosystems. Analyses of CLE exceedance used modelled NH_3 concentrations at a height of 1.5 m above ground. CL exceedance calculations were based on total N deposition: the LADD estimate of dry NH_3 deposition (see Section 2.4) plus the wet deposition of reduced N and the dry and wet deposition of oxidised N calculated using the UK FRAME model run for 2008 at a 1 km × 1 km resolution. The contribution of particulate ammonium (NH_4^+) to the dry deposition of reduced N is considered minor compared with NH_3 (e.g. Asman et al., 1998; Duyzer, 1994). Hallsworth et al. (2010) validated the FRAME model at a 1 km resolution for NH_3 concentrations and Dore et al. (2012) for NO_2 concentrations. Dore et al. (2007) validated the FRAME model for aerosol concentrations and wet deposition at 5 km resolution. FRAME gives three different deposition rates for each grid square: a) the average deposition, accounting for land cover mix in the grid square; b) the deposition to woodland in the square; c) the deposition to low semi-natural vegetation in the square. Deposition rates were applied depending on the land cover in each 25 m grid square. The CL exceedance was calculated for woodland, hedgerows, shrubs, moorland and rough grass by subtracting the CL of 10 kg N ha^{-1} yr^{-1} from the total N deposition. The applied CL is the lower limit of the range shown in Table 1 to protect the most sensitive species of the respective ecosystems. Although these CL values were used in this study, current science indicates that they may need to be revised further to adequately protect sensitive species (Payne et al., 2013).

3. Results and discussion

3.1. Spatial variability of measured NH_3 concentrations

The spatial variability of NH_3 concentrations in the landscape was large, with monthly NH_3 concentrations during 2008 varying from 0.2 to 42.5 μg m^{-3} between the measurement sites. Monthly coefficients of variation of replicate samplers was <24%, with values >15% only occurring at sites with monthly mean concentrations <1 μg m^{-3}. The spatial variability of the measured NH_3 concentrations is attributed to land use, as shown by classifying the

Table 1
Land cover categories of the study landscape, the associated ecosystem types with the corresponding critical loads for N deposition (UNECE, 2010).

Land cover category	Ecosystem type	Critical load [kg N ha^{-1} yr^{-1}]
Woodland, hedgerows	Broadleaved deciduous woodland	10–20
Shrubs	*Calluna* dominated wet heath (upland moorland)	10–20
Moorland, rough grass	Heath (*Juncus*) meadows and humid (*Nardus stricta*) swards	10–20

sites into three categories: a) "Background sites" are located away from agricultural NH_3 sources and have mean annual concentrations of <1 μg m^{-3}, b) "Field sites" are influenced by agricultural NH_3 sources such as grazing or fertiliser applications, but are >300 m away from large point sources, and c) "Poultry sites" within 300 m of large point sources, i.e. the poultry houses. Annual mean NH_3 concentrations in 2008 ranged between 0.40 and 22.9 μg NH_3 m^{-3} (Fig. 2) and generally increased from Background to Field to Poultry sites (Fig. 3). Two Field sites were exceptions with higher concentrations: Site 24 was close to an open cattle shed and an intensively used field, and site 25, which was only 320 m from a poultry house, i.e. just outside the distance for Poultry site classification.

The highest annual mean NH_3 concentrations were measured 70 m downwind (northeast) of a poultry house with an estimated NH_3 emission strength of 5900 kg N yr^{-1} (site 31). A measurement transect of three sites downwind of this house illustrates the concentration gradient with distance from large sources. Measured annual concentrations were 22.9 μg m^{-3}, 14.7 μg m^{-3} and 4.8 μg m^{-3} at distances of 70 m, 160 m and 900 m from the house (sites 31, 30, 23, respectively). Fig. 4 compares these results to concentration decreases with distance found by Fowler et al. (1998) and Pitcairn et al. (1998) for poultry houses emitting an estimated 4800 kg N yr^{-1} and 14,000 kg N yr^{-1}, respectively. All three studies were conducted in agricultural areas; however, the concentration decrease with distance in this study is more gradual, possibly due to high concentrations in the surrounding area resulting from the large number of emission hotspots.

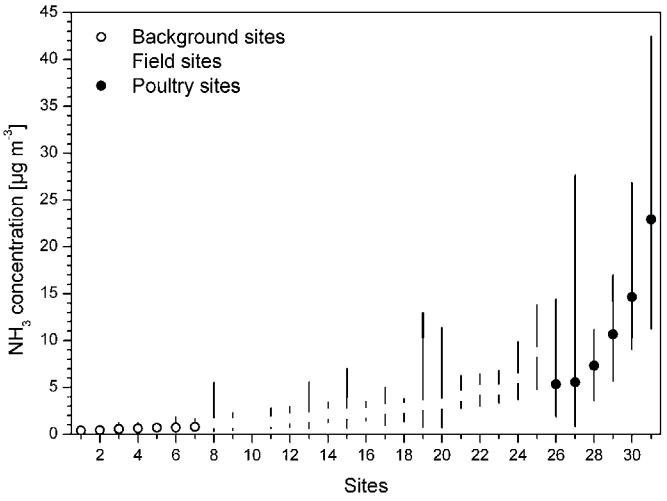

Fig. 3. Annual measured mean NH_3 concentrations and monthly minima and maxima in 2008 for Background sites (open circles), Field sites (grey circles) and Poultry sites (black circles). The site numbers ranked by site category and mean concentration are those shown in Fig. 2.

3.2. Temporal variability in measured NH_3 concentrations

A strong correlation was found between annual mean NH_3 concentrations of all sites in 2007 and 2008 (R^2 = 0.98, n = 31), using data from April to December for both years. This strong correlation indicates that the surrounding land use is the main driver of variation in the annual concentration. The ratio of monthly concentration maxima to annual mean concentrations can be used as an indicator of temporal variability on an intra-annual basis (Fig. 5). Most sites show a ratio below 3:1, which seems to represent a typical temporal variation about a mean of a relatively constant NH_3 concentration (e.g. Fig. 6a). This was also shown by Tang et al.

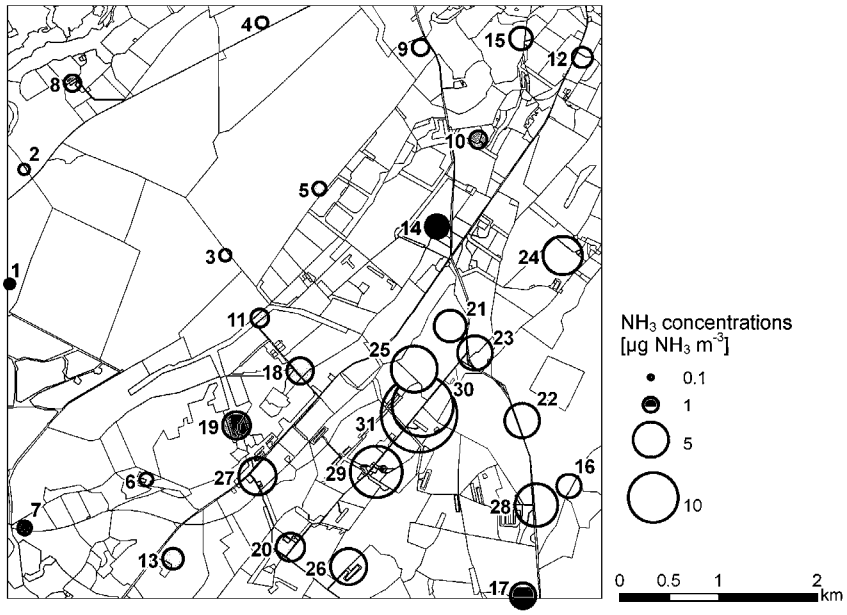

Fig. 2. Map of numbered measurement site locations showing annual mean NH_3 concentrations by proportionally sized circles. The sites are numbered in rank order of their mean NH_3 concentration within their site category.

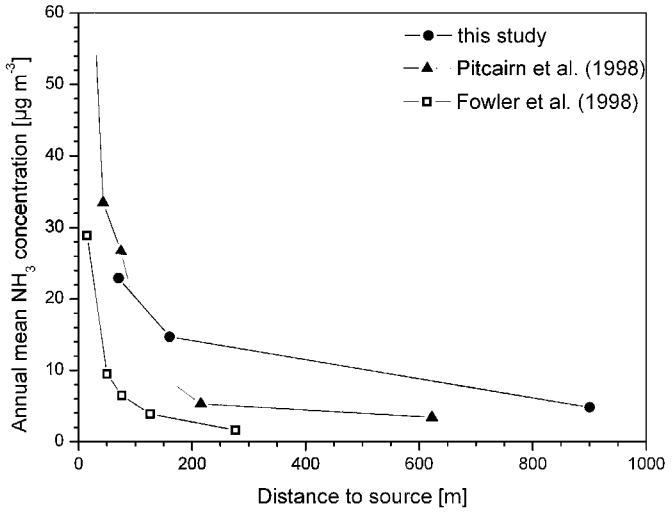

Fig. 4. Ammonia concentration decrease with distance from the source of this study (sites 31, 30, 23) compared with results of Pitcairn et al. (1998) and Fowler et al. (1998).

(2009) for the UK at the national scale. Monthly maximum concentrations of those sites with larger ratios (up to 5:1) occurred in spring or summer 2008. For example, site 27 (Fig. 6b) was located around 200 m south of four poultry houses, but it was also located close to a field where manure was applied in May 2008. Manure heaps and manure applications also accounted for monthly maxima at sites 8 (Fig. 6c), 13, 15, 19 and 20 (Fig. 6d).

3.3. Modelled atmospheric concentration and deposition

The LADD model was initially run using emission factors (EFs) from the UK inventory of NH_3 emissions and resulted in the general pattern of NH_3 concentrations being reproduced (Fig. 7, left). However there was a significant overestimation of concentration in the landscape, especially in the southeastern quarter. This overestimation was attributed to the emissions from six of the poultry houses (circled houses in Fig. 1) which contained ~3/4 million layers in cage systems. These houses had frequently cleaned belt-systems ($\geq$2 times week^{-1}). The EF for a UK average caged layer

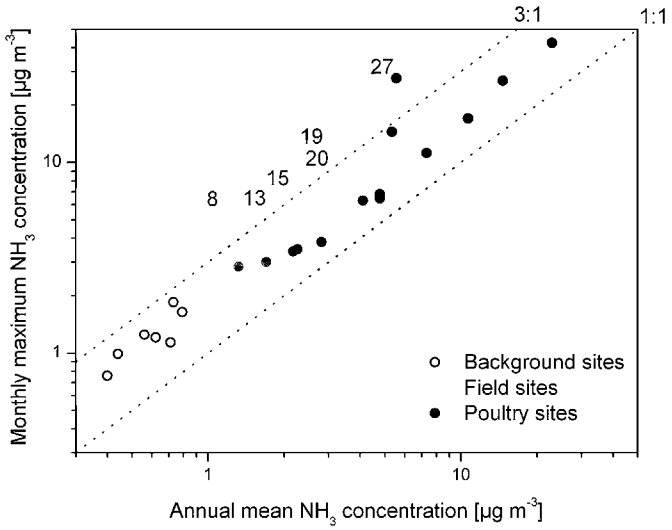

Fig. 5. Relationship between monthly maximum and annual mean NH_3 concentrations in 2008 for Background sites (open circles), Field sites (grey circles) and Poultry sites (black circles). Site numbers are shown for sites with ratios of the monthly maximum to the annual mean higher than 3:1.

is calculated assuming that 40% of these birds are housed in deep-pit houses and 60% in belt-system houses with a cleaning frequency of $\leq$1 times week^{-1} (Misselbrook et al., 2009). Belt-systems with less frequent cleaning (EF = 0.092 kg NH_3–N bird^{-1} yr^{-1}) are considered to reduce emissions by 56% compared to deep-pit systems (EF = 0.164 kg NH_3–N bird^{-1} yr^{-1}), resulting in an average UK caged layer EF of 0.121 kg NH_3–N bird^{-1} yr^{-1} (Misselbrook et al., 2009). The European Commission (2003) reported an EF of 0.029 kg NH_3–N bird^{-1} yr^{-1} for frequently cleaned belt-systems, more than four times lower than that used in the UK NH_3 inventory for the average caged layer. LADD runs were repeated using this EF for the six poultry houses concerned and modelled concentrations decreased considerably (Fig. 7, right), and matched measured concentrations more closely.

Fig. 8 shows a scatter plot between modelled and measured concentrations and Table 2 summarises the statistical metrics for the model run with system specific EFs applied for the poultry houses instead of UK average EFs. Overall, model performance is evaluated as acceptable, as the FAC2 and VG metrics indicate acceptable model performance when compared with measurements at all sites. However, the MG is lower than recommended for acceptable model performance, reflecting a systematic overestimation by the model, which is apparent at all distances from sources (Fig. 8).

Recent work by Theobald et al. (2012) suggests that LADD overestimates concentrations around elevated sources (>5 m) with high exit velocities, as LADD does not include treatment of plume rise after leaving the source. However, poultry houses in this study area predominantly have emission heights of 4–5 m, and most vents are located on the building walls, i.e. most plumes are not expected to exit vertically. Thus this is unlikely to explain the differences shown in Fig. 8. For other situations with ground and building emission sources, Theobald et al. (2012) reported acceptable agreement between LADD and measured concentrations.

3.4. Model calibration

In order to use modelled concentrations and deposition fluxes for risk assessment of environmental impacts, the systematic overestimation was addressed by calibrating the modelled against the measured concentrations. Modelled concentrations were corrected by the slope of the regression between measured and modelled results ($[NH_3]_{meas} = 0.49\ [NH_3]_{model} + 0.15$, $R^2 = 0.90$). The intercept was not statistically significant, providing the simplified relationship $[NH_3]_{meas} = 0.49\ [NH_3]_{model}$ ($R^2 = 0.90$) to calibrate modelled concentrations (effectively represented by the 2:1 function in Fig. 8).

The calibrated model NH_3 concentrations range from 0.3 to 77.9 µg m^{-3} within the study landscape (Fig. 9). These results provide the basis to use the model for assessing the risk of environmental impacts in the study landscape at high spatial resolution.

3.5. Risk assessment of environmental impacts

The risk assessment presented is based on comparison of the NH_3 concentrations with the CLE and N deposition with the CL focussing on the extent of CLE and CL exceedance. For this purpose, the high resolution 25 m calibrated model output data of NH_3 concentration and dry NH_3 deposition were supplemented with the 1 km national deposition estimates for oxidised N and wet-deposited reduced N for the study domain. In addition, landscape scale and national scale assessments were compared to investigate fitness-for purpose at the different spatial resolutions.

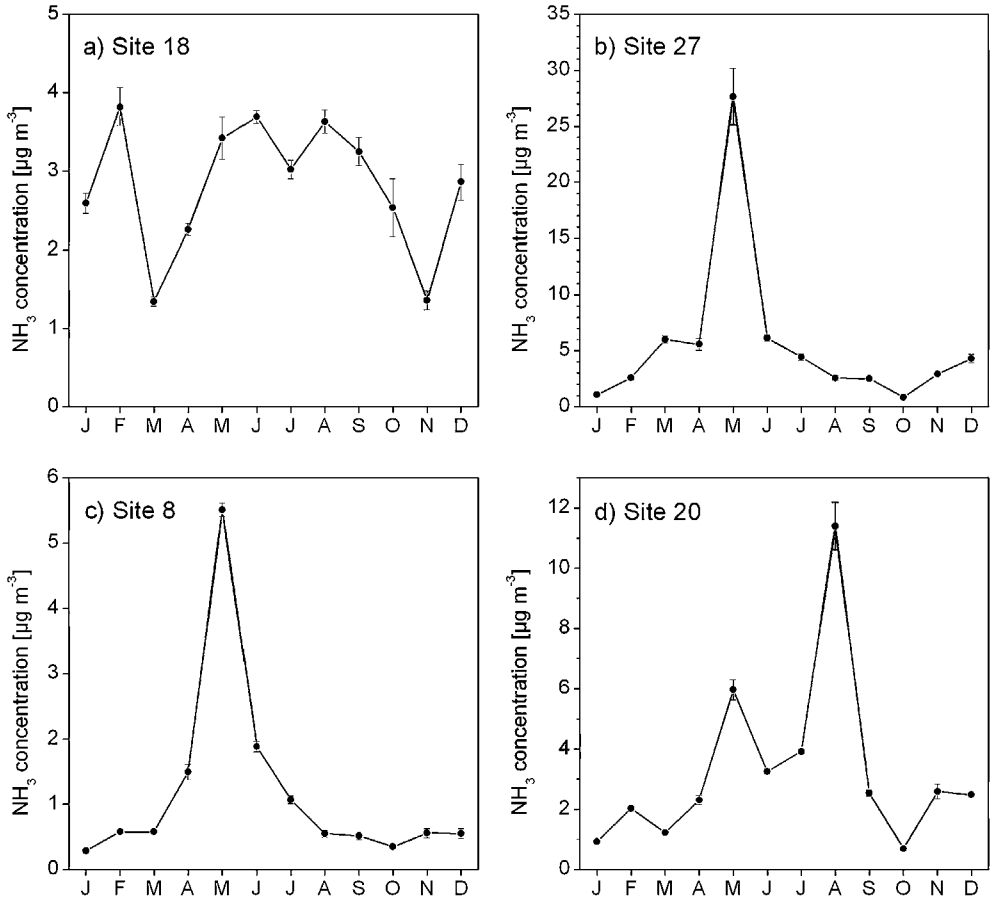

Fig. 6. Temporal variation of monthly NH_3 concentrations (±2 standard deviations) during 2008 at four sites: a) Site 18 with a ratio of max/mean below 3:1 and b), c) and d) showing sites with ratios higher than 3:1.

3.5.1. Concentrations and critical level (CLE) exceedance

For sensitive vegetation (regardless of habitat type), i.e. lichens and bryophytes, the long term CLE for NH_3 of 1 µg m^{-3} is exceeded in 60% of the landscape (Fig. 10). Moorland habitats are naturally low N ecosystems and sensitive to NH_3. Within the study area, the CLE is exceeded for 8% of the moorland areas. Such ecosystems could thus be expected to show long term effects of local NH_3 sources. Although this affects a considerable area (39 ha moorland), it is still a relatively modest fraction considering the extremely high emission fluxes in the vicinity. This is due to most of the moorland

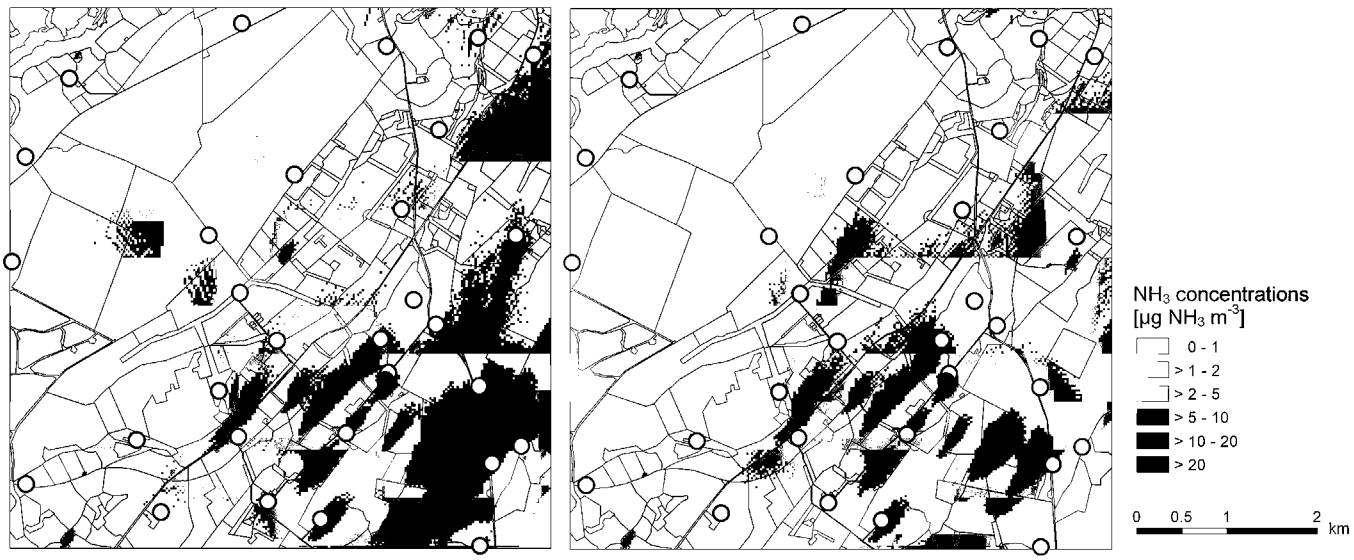

Fig. 7. Measured (circles) and modelled (background colours) NH_3 concentrations within the landscape. Left map: UK inventory emission factors were applied to all NH_3 sources; Right map: the European Commission (2003) EF for frequently cleaned belt-systems was applied to the six poultry houses that had that system.

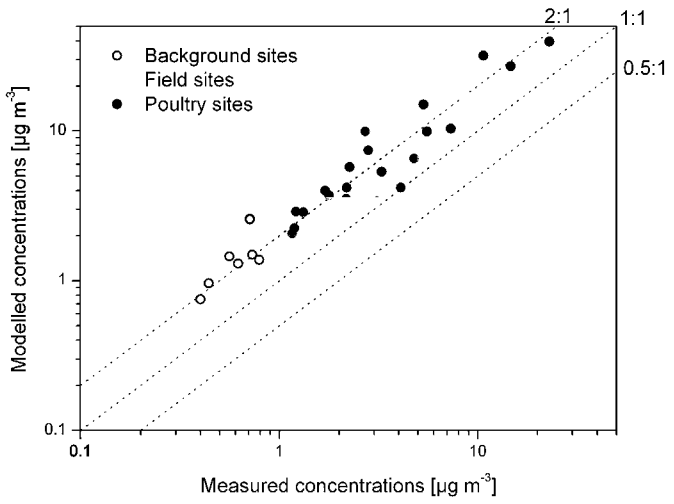

Fig. 8. Relationship between modelled and measured NH_3 concentrations of Background sites (open circles), Field sites (grey circles) and Poultry sites (black circles) on logarithmic axes.

Table 2
Statistical metrics of model performance comparing measured and modelled NH_3 concentrations for all sites and by separate site categories (see Section 3.1 for category definition).

	Target performance	All sites	Background sites	Field sites	Poultry sites
FAC2 (%)	≥50.0	51.6	28.6	55.6	66.7
R	–	0.95	0.64	0.84	0.89
MG	0.7–1.3	0.50	0.45	0.52	0.50
VG	<3.3	1.77	2.03	1.68	1.76

in the study area being located northwest of the poultry houses, in a region with frequent southwesterly winds and low NH_3 background concentration (0.40 µg m^{-3} at site 1).

The CLE of 3 µg m^{-3} for higher plants is exceeded in 25% of the landscape. Most of this area is agricultural land: 81% is grass or arable land, and the risk of species composition change is not an issue in arable crops or improved grassland with already substantial fertiliser N input. However, semi-natural vegetation and woodland areas in the landscape with NH_3 concentrations >3 µg m^{-3} are at risk, with 7% of such habitats showing an exceedance of the CLE. However, these habitats exceeding the CLE of 3 µg m^{-3} are restricted to relatively small patches within the agricultural area.

These datasets were aggregated from a 25 m resolution to 1 km × 1 km for comparison with concentrations modelled by the UK scale FRAME model at a 1 km resolution using national emission factors (Table 3). FRAME predicts exceedances of the 1 µg m^{-3} CLE for the whole landscape, and conversely no exceedances for the 3 µg m^{-3} CLE. In other words, at the coarser resolution, the impact to the sensitive moorland area northwest of the emission hotspots is overestimated, while the impact downwind of the hotspots is substantially underestimated. Thus, FRAME smoothes out the full spatial variability of NH_3 concentrations at 1 km resolution to a large degree. By contrast, LADD NH_3 concentrations aggregated to a 1 km resolution capture a much higher level of spatial heterogeneity in CLE exceedances (Table 3, Figs. 11 and 12). This suggests that the smoothing out of NH_3 concentrations across a landscape in a national scale model such as FRAME is largely due to coarser scale input data, i.e. during the emission inventory processing. FRAME uses spatial patterns of UK NH_3 emissions calculated in the AENEID model (Dragosits et al., 1998; Hellsten et al., 2008). The model combines parish-level farm statistics with weighted component NH_3 sources according to land cover at 1 km level. Hallsworth et al. (2010) have shown that this approach provides encouraging agreement with NH_3 concentrations modelled at the national scale (model-measurement comparison: $R^2 = 0.83$), due to 1 km model simulations more effectively separating source (agricultural) areas from sink (semi-natural/nature reserve) areas than the 5 km model.

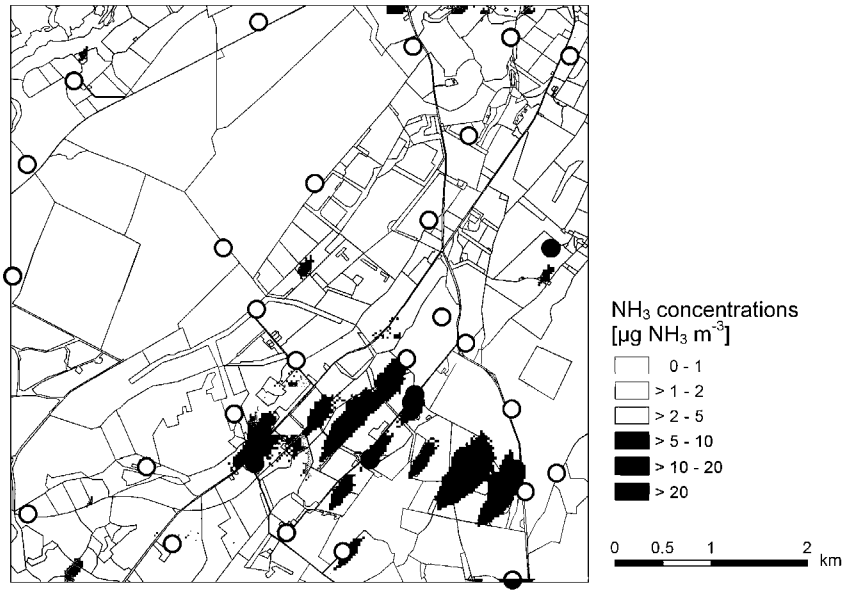

Fig. 9. Measured (circles) and calibrated modelled (background colours) NH_3 concentrations within the landscape. For all NH_3 sources, except for six frequently cleaned poultry houses, average emission factors from the UK inventory were used as model input.

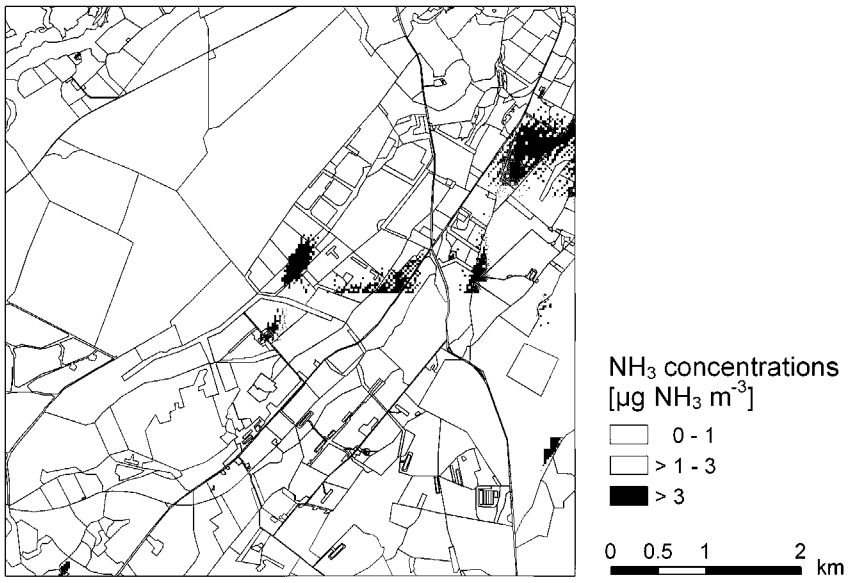

Fig. 10. Modelled NH_3 concentrations (calibrated) within the study landscape. In white areas, vegetation is not expected to be at risk through NH_3 concentrations. In light and dark grey areas, NH_3 concentrations pose a risk to sensitive vegetation, such as lichens and bryophytes. In dark grey areas, all plants of semi-natural ecosystems are at risk.

The present study illustrates the limitation of applying a national scale approach at high resolution to a specific landscape, as UK emission mapping is based on general suitability of different land classes for agriculture, but does not include detailed mapping of agricultural point source emissions.

A scenario with all poultry emissions removed was tested for CLE exceedance. In this no-poultry scenario only 12% of the study area exceeded the 1 µg m^{-3} CLE, and 0.2% exceeded the 3 µg m^{-3} CLE, compared with 60% and 25%, respectively, when poultry house emissions were included. This highlights the large contribution of emission hotspots to atmospheric NH_3 concentrations in mixed landscapes such as this.

3.5.2. Deposition and critical load (CL) exceedance

Modelled dry deposition of NH_3 within the landscape has a high spatial variability ranging from 0.1 to 1200 kg NH_3–N ha^{-1} yr^{-1}, with a value >1000 kg NH_3–N ha^{-1} yr^{-1} occurring in only a single 25 m × 25 m grid square between closely located poultry houses. Such high dry deposition values can be considered theoretical estimates, as the deposition rate may be expected to be reduced close to large sources due to saturation of the absorbing surfaces (Jones et al., 2007). In most cases, the deposition decreases to <100 kg NH_3–N ha^{-1} yr^{-1} within 100 m distance from a source, depending on the absorbing surfaces. To illustrate the importance of capturing the spatial variability, the deposition flux to coniferous woodland downwind of a poultry house was compared with estimates by FRAME (circled area in Fig. 13). The

Table 3
Comparison of the range of modelled NH_3 concentrations within the study landscape and the percentage of CLE exceedance at different resolutions: LADD (25 m, 1 km) and FRAME (1 km).

	LADD – 25 m	LADD – 1 km	FRAME – 1 km
Min (µg m^{-3})	0.3	0.4	1.1
Max (µg m^{-3})	77.9	10.7	2.9
Mean (µg m^{-3})	2.6	2.6	1.9
% CLE exceedance 1 µg m^{-3}	60	64	100
% CLE exceedance 3 µg m^{-3}	25	31	0

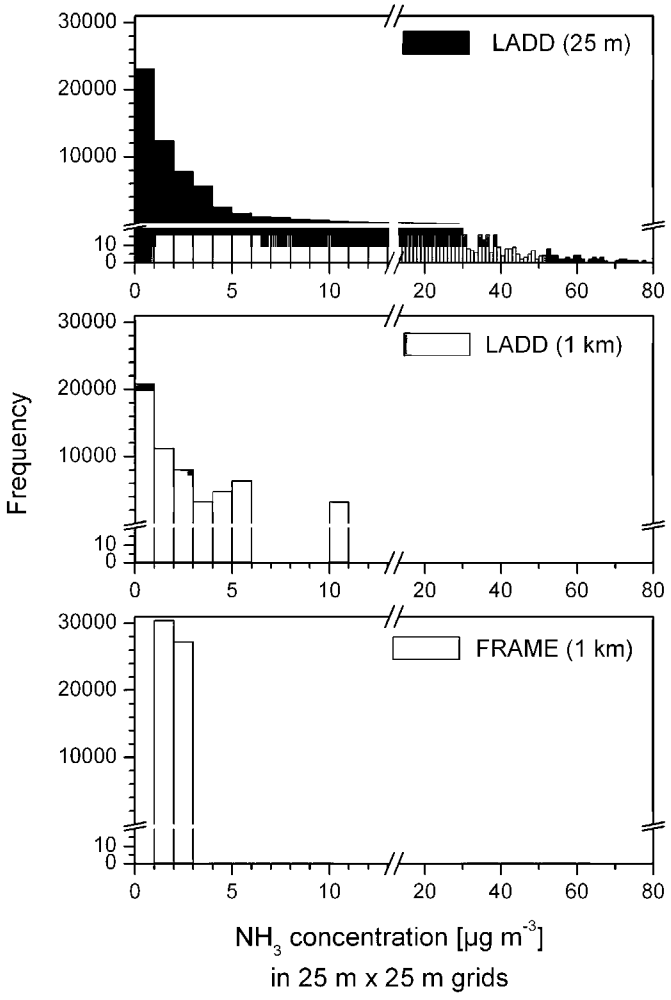

Fig. 11. Histograms of modelled NH_3 concentrations in 25 m × 25 m grids in the landscape comparing results at different resolutions: LADD (25 m, 1 km) and FRAME (1 km).

Fig. 12. NH_3 concentrations at a 1 km resolution within the study landscape, using FRAME (left map) and LADD (right map).

woodland of 6.5 ha is situated between 150 m and 500 m from the house. The NH_3 dry deposition flux to the woodland modelled by LADD varies spatially between 31 and 172 kg N ha^{-1} yr^{-1} and amounts to a total of 394 kg N yr^{-1}. This is equivalent to 6.7% of the NH_3 emitted from the poultry house near site 31, though other poultry houses would have also contributed to this total. FRAME at 1 km estimates a woodland specific dry deposition flux to this area of 10.8–11.9 kg N ha^{-1} yr^{-1} (total NH_x dry deposition of 74 kg N yr^{-1}). This illustrates how FRAME underestimates the impact of NH_x dry deposition in the immediate vicinity of sources, compared with LADD.

Total N deposition (LADD NH_3 dry deposition + FRAME NH_x wet & NO_y deposition) ranges from 5.6 to 1206 kg N ha^{-1} yr^{-1} (Fig. 13). The CL only applies to the land cover categories woodland, hedgerows, shrubs, moorland and rough grass in the landscape, i.e. CL exceedances were calculated only for these categories, which cover 38% of the study area. In 34% of this area (or 13% of the total landscape area) the CL is exceeded, on average by 17.6 kg N ha^{-1} yr^{-1}, with a median CL exceedance of 6.5 kg N ha^{-1} yr^{-1} (Fig. 14 and Table 4).

When combining the land cover specific 1 km FRAME results with the 25 m grid resolution land cover data (see Section 2.6), FRAME predicts a CL exceedance in 51% of the area to which a CL applies, compared with 34% simulated by LADD (Table 5). Due to FRAME not fully capturing the spatial variability of NH_3 dry deposition, areas exceeding CL in the whole study landscape are overestimated whereas the extent of CL exceedance in areas close to sources is underestimated (Fig. 14).

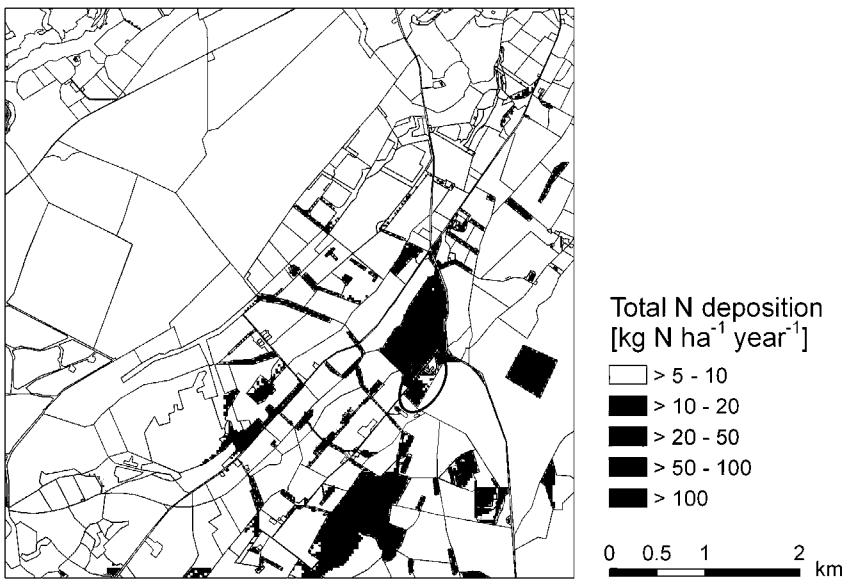

Fig. 13. Total N deposition calculated by combining dry deposition of NH_3 simulated by LADD (calibrated) with the remaining components of N deposition from FRAME. The circled area shows a patch of woodland analysed in more detail.

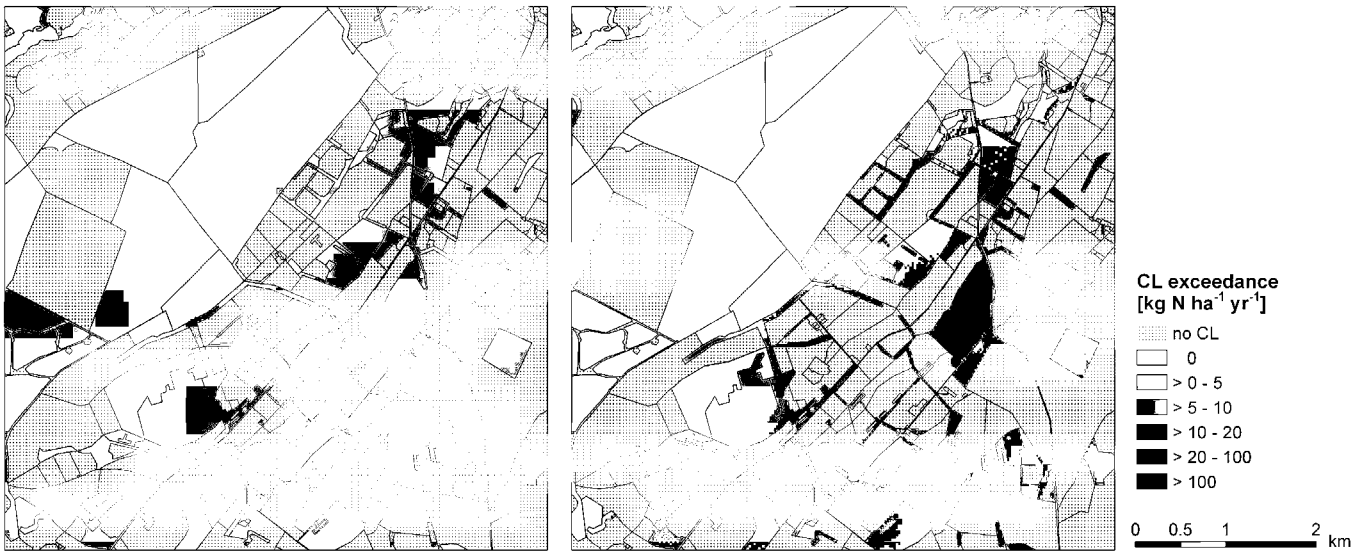

Fig. 14. Critical load (CL) exceedance calculated using land cover data and CL estimates at 25 m resolution and land cover specific atmospheric deposition estimates at differing spatial resolutions. Left: 1 km N deposition modelled by FRAME; Right: combining 25 m NH_3 dry deposition simulated by LADD (calibrated) with remaining components of N deposition from FRAME.

Table 4
Land cover specific statistics[a] for critical load (CL) exceedance (kg N ha^{-1} yr^{-1}).

	Woodland	Shrubs	Rough grass	Moorland
Mean	20.1	21.6	11.6	1.9
Median	7.4	17.6	2.7	0.7
Maximum	1195.6	401.9	406.5	10.5
% exceeding CL	74.2	97.0	28.0	1.7

[a] Land cover category hedgerows covered only a very small area and was therefore not considered for these statistics.

Table 5
Comparison of CL exceedances (kg N ha^{-1} yr^{-1}) within the study landscape between LADD (25 m resolution) and FRAME (1 km resolution).

	LADD	FRAME
Mean	17.6	3.2
Median	6.5	2.4
Maximum	1195.6	10.8
% exceeding CL	34	51

4. Conclusions

A detailed landscape inventory of all farm activities in the study year 2008 provided data to estimate NH_3 emissions at 25 m resolution. This is essential for studying the actual spatial variability of NH_3 at the landscape scale. The combination of a large number of long term NH_3 concentration measurements across the landscape and the high resolution model output allowed a spatially precise assessment of NH_3 concentrations, which was applied to estimate NH_3 dry deposition. Measured and modelled NH_3 were highly correlated ($R^2 = 0.90$), but model estimates needed to be calibrated by approximately a factor of two to the measurements for environmental risk assessment. This highlights the importance of always including verification measurements in such an assessment. This is also highlighted by Theobald et al. (2012), who showed that the performance of models such as LADD, ADMS and AERMOD, can vary between study sites, for example depending on specific meteorological and emission source characteristics. For robust risk assessment of environmental impacts, models therefore should be appropriately verified with measurements at multiple locations across the study area.

In the present study area, it was also found that standard national emission factors (EFs) were not appropriate for all main NH_3 sources. Ammonia EFs for several poultry houses had to be adjusted to account for the specific manure management practices of frequent litter removal, which resulted in a 75% decrease in emissions compared to the UK average EFs. Thus, for the environmental impact assessment of large livestock houses, it is important to use EFs appropriate to local husbandry systems and manure management.

In this study area, frequent southwesterly winds cause most of the poultry house emissions to disperse to the northeast. As the most sensitive ecosystems in the study area are located northwest of the poultry houses, only a relatively small area is affected by the nearby poultry emissions, despite total poultry NH_3 emissions exceeding 100 t N yr^{-1}, in addition to ~10 t N yr^{-1} of NH_3 emissions from other agricultural sources in the landscape. The ecosystems most at risk from high NH_3 concentrations are patches of woodland, shrubs and rough grass situated within the agricultural area downwind of the sources. Impact assessment using the critical level (CLE) approach suggested that 8% of the semi-natural moorland may be adversely affected by NH_3 concentrations above 1 μg m^{-3} (= long term CLE for lichens and bryophytes). By comparison, only 2% of the moorland area is under threat from critical load (CL) exceedance. This relatively small difference between two complementary environmental indicator approaches shows that the present value of the NH_3 CLE is in reasonably close agreement with the CL values in this upland landscape.

The comparison of the UK national model FRAME at a 1 km resolution with the 25 m resolution LADD estimates showed that FRAME did not capture the full spatial variability of NH_3 within the study landscape. Furthermore, a comparison of LADD NH_3 concentrations averaged to 1 km with FRAME 1 km concentrations showed the much higher potential to represent the spatial heterogeneity of NH_3 in the landscape framework. While the 1 km resolution version from FRAME performs well at the national scale (Hallsworth et al., 2010), this comparison emphasises the need for

high resolution emission data obtained at a farm and field level for assessments of environmental impacts from NH_3.

This study highlights the importance of the spatial arrangement of NH_3 sources and sinks within a landscape that is the cause of fine scale heterogeneity in NH_3 concentrations and N deposition and in the resulting environmental risks. In the study landscape, most sensitive ecosystems are located upwind of the large NH_3 sources nearby and thus are considered to be at relatively modest environmental risk according to current values of CLEs and CLs. This shows how landscape planning could be used to reduce the impact of intensive agriculture on sensitive ecosystems: Careful planning of the location of the farm point and area sources, considering both distance and the direction in relation to prevailing winds, provides a practical way of avoiding adverse impacts on nearby semi-natural areas.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2013.04.014.

References

Asman, W.A.H., Sutton, M.A., Schjørring, J.K., 1998. Ammonia: emission, atmospheric transport and deposition. New Phytologist 139, 27–48.

Bouwman, A.F., Lee, D.S., Asman, W.A.H., Dentener, F.J., Van Der Hoek, K.W., Olivier, J.G.J., 1997. A global high-resolution emission inventory for ammonia. Global Biogeochemical Cycles 11, 561–587.

Burkhardt, J., Sutton, M.A., Milford, C., Storeton-West, R.L., Fowler, D., 1998. Ammonia concentrations at a site in southern Scotland from 2 yr of continuous measurements. Atmospheric Environment 32, 325–331.

Cape, J.N., van der Eerden, L.J., Sheppard, L.J., Leith, I.D., Sutton, M.A., 2009a. Evidence for changing the critical level for ammonia. Environmental Pollution 157, 1033–1037.

Cape, J.N., van der Eerden, L.J., Sheppard, L.J., Leith, I.D., Sutton, M.A., 2009b. Reassessment of critical levels for atmospheric ammonia. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 15–40.

Cellier, P., Durand, P., Hutchings, N., Dragosits, U., Theobald, M.R., Drouet, J.-L., Oenema, O., Bleeker, A., Breuer, L., Dalgaard, T., Duretz, S., Kros, J., Loubet, B., Olesen, J.E., Merot, P., Viaud, V., de Vries, W., Sutton, M.A., 2011. Nitrogen flows and fate in rural landscapes. In: Sutton, M.A., et al. (Eds.), The European Nitrogen Assessment – Sources, Effects and Policy Perspectives. Cambridge University Press, Cambridge, pp. 229–248.

Cellier, P., Theobald, M.R., Asman, W., Bealey, W., Bittman, S., Dragosits, U., Fudala, J., Jones, M., Lofstrom, P., Loubet, B., Misselbrook, T., Rihm, B., Smith, K., Strizik, M., van der Hoek, K., van Jaarsveld, H., Walker, J., Zelinger, Z., 2009. Assessment methods for ammonia hot-spots. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 391–407.

Chang, J.C., Hanna, S.R., 2004. Air quality model performance evaluation. Meteorology and Atmospheric Physics 87, 167–196.

Defra, 2010. Department for Environmental Food and Rural Affairs: Fertiliser Manual (RB209), eighth ed. TSO (The Stationary Office), Norwich, UK.

Dore, A.J., Kryza, M., Hall, J.R., Hallsworth, S., Keller, V.J.D., Vieno, M., Sutton, M.A., 2012. The influence of model grid resolution on estimation of national scale nitrogen deposition and exceedance of critical loads. Biogeosciences 9, 1597–1609.

Dore, A.J., Vieno, M., Tang, Y.S., Dragosits, U., Dosio, A., Weston, K.J., Sutton, M.A., 2007. Modelling the atmospheric transport and deposition of sulphur and nitrogen over the United Kingdom and assessment of the influence of SO_2 emissions from international shipping. Atmospheric Environment 41, 2355–2367.

Dragosits, U., Sutton, M.A., Place, C.J., Bayley, A.A., 1998. Modelling the spatial distribution of agricultural ammonia emissions in the UK. Environmental Pollution 102, 195–203.

Dragosits, U., Theobald, M.R., Place, C.J., ApSimon, H.M., Sutton, M.A., 2006. The potential for spatial planning at the landscape level to mitigate the effects of atmospheric ammonia deposition. Environmental Science & Policy 9, 626–638.

Dragosits, U., Theobald, M.R., Place, C.J., Lord, E., Webb, J., Hill, J., ApSimon, H.M., Sutton, M.A., 2002. Ammonia emission, deposition and impact assessment at the field scale: a case study of sub-grid spatial variability. Environmental Pollution 117, 147–158.

Duyzer, J., 1994. Dry deposition of ammonia and ammonium aerosols over heathland. Journal of Geophysical Research 99, 18757–18763.

European Commission, 2003. Integrated Pollution Prevention and Control (IPPC). Reference Document on Best Available Techniques for Intensive Rearing of Poultry and Pigs (BREF-ILF), Seville, Spain.

Fangmeier, A., Hadwiger-Fangmeier, A., Van der Eerden, L., Jäger, H.-J., 1994. Effects of atmospheric ammonia on vegetation – a review. Environmental Pollution 86, 43–82.

Forman, R.T.T., Godron, M., 1981. Patches and structural components for a landscape ecology. Bioscience 31, 733–740.

Fowler, D., Pitcairn, C.E.R., Sutton, M.A., Flechard, C., Loubet, B., Coyle, M., Munro, R.C., 1998. The mass budget of atmospheric ammonia in woodland within 1 km of livestock buildings. Environmental Pollution 102, 343–348.

Frati, L., Santoni, S., Nicolardi, V., Gaggi, C., Brunialti, G., Guttova, A., Gaudino, S., Pati, A., Pirintsos, S.A., Loppi, S., 2007. Lichen biomonitoring of ammonia emission and nitrogen deposition around a pig stockfarm. Environmental Pollution 146, 311–316.

Hallsworth, S., Dore, A.J., Bealey, W.I., Dragosits, U., Vieno, M., Hellsten, S., Tang, Y.S., Sutton, M.A., 2010. The role of indicator choice in quantifying the threat of atmospheric ammonia to the 'Natura 2000' network. Environmental Science & Policy 13, 671–687.

Hanna, S.R., Chang, J.C., 2010. Setting acceptance criteria for air quality models. In: Proceedings of the International Technical Meeting on Air Pollution Modelling and its Application, Turin, Italy.

Hellsten, S., Dragosits, U., Place, C.J., Vieno, M., Dore, A.J., Misselbrook, T.H., Tang, Y.S., Sutton, M.A., 2008. Modelling the spatial distribution of ammonia emissions in the UK. Environmental Pollution 154, 370–379.

Hill, J., 1998. Applications of Computational Modelling to Ammonia Dispersion from Agricultural Sources. Ph.D. thesis. Imperial College, Centre for Environmental Technology, University of London, London, UK.

Jones, M.R., Leith, I.D., Fowler, D., Raven, J.A., Sutton, M.A., Nemitz, E., Cape, J.N., Sheppard, L.J., Smith, R.I., Theobald, M.R., 2007. Concentration-dependent NH_3 deposition processes for mixed moorland semi-natural vegetation. Atmospheric Environment 41, 2049–2060.

Krupa, S.V., 2003. Effects of atmospheric ammonia (NH_3) on terrestrial vegetation: a review. Environmental Pollution 124, 179–221.

Loubet, B., Asman, W.A.H., Theobald, M.R., Hertel, O., Tang, Y.S., Robin, P., Hassouna, M., Dammgen, U., Genermont, S., Cellier, P., Sutton, M.A., 2009. Ammonia deposition near hot spots: processes, models and monitoring methods. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 205–267.

Matejko, M., Dore, A.J., Hall, J., Dore, C.J., Blas, M., Kryza, M., Smith, R., Fowler, D., 2009. The influence of long term trends in pollutant emissions on deposition of sulphur and nitrogen and exceedance of critical loads in the United Kingdom. Environmental Science & Policy 12, 882–896.

Misselbrook, T.H., Chadwick, D.R., Gilhespy, S.L., Chambers, B.J., Smith, K.A., Williams, J., Dragosits, U., 2009. Inventory of Ammonia Emissions from UK Agriculture 2008 (Defra Contract AC0112). North Wyke Research, Devon, UK.

Misselbrook, T.H., Van Der Weerden, T.J., Pain, B.F., Jarvis, S.C., Chambers, B.J., Smith, K.A., Phillips, V.R., Demmers, T.G.M., 2000. Ammonia emission factors for UK agriculture. Atmospheric Environment 34, 871–880.

Payne, R.J., Dise, N.B., Stevens, C.J., Gowing, D.J., Partners, B., 2013. Impact of nitrogen deposition at the species level. Proceedings of the National Academy of Sciences 110, 984–987.

Pitcairn, C.E.R., Leith, I.D., Sheppard, L.J., Sutton, M.A., Fowler, D., Munro, R.C., Tang, S., Wilson, D., 1998. The relationship between nitrogen deposition, species composition and foliar nitrogen concentrations in woodland flora in the vicinity of livestock farms. Environmental Pollution 102, 41–48.

Pitcairn, C.E.R., Leith, I.D., van Dijk, N., Sheppard, L.J., Sutton, M.A., Fowler, D., 2009. The application of transects to assess the effects of ammonia on woodland groundflora. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 59–69.

Pitcairn, C.E.R., Skiba, U.M., Sutton, M.A., Fowler, D., Munro, R., Kennedy, V., 2002. Defining the spatial impacts of poultry farm ammonia emissions on species composition of adjacent woodland groundflora using Ellenberg Nitrogen Index, nitrous oxide and nitric oxide emissions and foliar nitrogen as marker variables. Environmental Pollution 119, 9–21.

Posthumus, A.C., 1988. Critical levels for effects of ammonia and ammonium. In: Proceedings of the Bad Harzburg Workshop. Umweltbundesamt, Berlin, pp. 117–127.

Sutton, M.A., Milford, C., Dragosits, U., Place, C.J., Singles, R.J., Smith, R.I., Pitcairn, C.E.R., Fowler, D., Hill, J., ApSimon, H.M., Ross, C., Hill, R., Jarvis, S.C., Pain, B.F., Phillips, V.C., Harrison, R., Moss, D., Webb, J., Espenhahn, S.E., Lee, D.S.,

Hornung, M., Ullyett, J., Bull, K.R., Emmett, B.A., Lowe, J., Wyers, G.P., 1998. Dispersion, deposition and impacts of atmospheric ammonia: quantifying local budgets and spatial variability. Environmental Pollution 102, 349–361.

Sutton, M.A., Miners, B., Tang, Y.S., Milford, C., Wyers, G.P., Duyzer, J.H., Fowler, D., 2001a. Comparison of low cost measurement techniques for long-term monitoring of atmospheric ammonia. Journal of Environmental Monitoring 3, 446–453.

Sutton, M.A., Tang, Y.S., Dragosits, U., Fournier, N., Dore, A.J., Smith, R.I., Weston, K.J., Fowler, D., 2001b. A spatial analysis of atmospheric ammonia and ammonium in the UK. The Scientific World Journal 1, 275–286.

Sutton, M.A., Nemitz, E., Erisman, J.W., Beier, C., Bahl, K.B., Cellier, P., de Vries, W., Cotrufo, F., Skiba, U., Di Marco, C., Jones, S., Laville, P., Soussana, J.F., Loubet, B., Twigg, M., Famulari, D., Whitehead, J., Gallagher, M.W., Neftel, A., Flechard, C.R., Herrmann, B., Calanca, P.L., Schjoerring, J.K., Daemmgen, U., Horvath, L., Tang, Y.S., Emmett, B.A., Tietema, A., Penuelas, J., Kesik, M., Brueggemann, N., Pilegaard, K., Vesala, T., Campbell, C.L., Olesen, J.E., Dragosits, U., Theobald, M.R., Levy, P., Mobbs, D.C., Milne, R., Viovy, N., Vuichard, N., Smith, J.U., Smith, P., Bergamaschi, P., Fowler, D., Reis, S., 2007. Challenges in quantifying biosphere-atmosphere exchange of nitrogen species. Environmental Pollution 150, 125–139.

Sutton, M.A., Reis, S., Baker, S.M.H., 2009a. Synthesis and summary for policy makers. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 445–454.

Sutton, M.A., Sheppard, L.J., Fowler, D., 2009b. Potential for the further development and application of critical levels to assess the environmental impacts of ammonia. In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 41–48.

Tang, Y.S., Cape, J.N., Sutton, M.A., 2001. Development and types of passive samplers for monitoring atmospheric NO_2 and NH_3 concentrations. The Scientific World Journal 1, 513–529.

Tang, Y.S., Dragosits, U., van Dijk, N., Love, L., Simmons, I., Sutton, M.A., 2009. Assessment of ammonia and ammonium trends and relationship to critical levels in the UK National Ammonia Monitoring Network (NAMN). In: Sutton, M.A., Reis, S., Baker, S.M.H. (Eds.), Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts. Springer, Dordrecht, pp. 187–194.

Theobald, M.R., Bealey, W.J., Tang, Y.S., Vallejo, A., Sutton, M.A., 2009. A simple model for screening the local impacts of atmospheric ammonia. Science of the Total Environment 407, 6024–6033.

Theobald, M.R., Løfstrøm, P., Walker, J., Andersen, H.V., Pedersen, P., Vallejo, A., Sutton, M.A., 2012. An intercomparison of models used to simulate the short-range atmospheric dispersion of agricultural ammonia emissions. Environmental Modelling & Software 37, 90–102.

Theobald, M.R., Milford, C., Hargreaves, K.J., Sheppard, L.J., Nemitz, E., Tang, Y.S., Phillips, V.R., Sneath, R., McCartney, L., Harvey, F.J., Leith, I.D., Cape, J.N., Fowler, D., Sutton, M.A., 2001. Potential for ammonia recapture by farm woodlands: design and application of a new experimental facility. The Scientific World 1, 791–801.

UNECE, 2007. Review of the 1999 Gothenburg Protocol: Report on the Workshop on Atmospheric Ammonia: Detecting Emission Changes and Environmental Impacts. In: Executive Body for the Convention on Long-range Transboundary Air Pollution, Working Group on Strategies and Review. 39th Session, 18–20 April 2007, Geneva, Switzerland (ECE/EB.AIR/WG.5/2007/3).

UNECE, 2010. Empirical Critical Loads and Dose-response Relationships. In: Convention on Long-range Transboundary Air Pollution, Working Group on Effects. 29th Session, 22–24 September 2010, Geneva, Switzerland. http://www.unece.org/env/documents/2010/eb/wge/ece.eb.air.wg.1.2010.14.e.pdf (February 2011).

Van der Hoek, K.W., 1998. Estimating ammonia emission factors in Europe: summary of the work of the UNECE ammonia expert panel. Atmospheric Environment 32, 315–316.

Wyers, G.P., Otjes, R.P., Slanina, J., 1993. A continuous-flow denuder for the measurement of ambient concentrations and surface-exchange fluxes of ammonia. Atmospheric Environment Part A-General Topics 27, 2085–2090.